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## Interpretation of variations in fine, coarse and black smoke particulate matter concentrations in a northern European city

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## Abstract

The PM<sub>2.5</sub>, PM<sub>coarse</sub> and Black Smoke (BS) particle metrics broadly reflect different source contributions to PM<sub>10</sub>. The aim of this study was to generate data for PM<sub>2.5</sub> at an urban background site in the UK, and to use the daily collocated measurement of PM<sub>2.5</sub>, PM<sub>10</sub> (and hence PM<sub>coarse</sub>) and BS to yield insight into source influences on particulate matter for input to developing PM air quality policy. Mean daily PM<sub>10</sub>, PM<sub>2.5</sub> and BS for a year of measurement in Edinburgh were 15.5, 8.5 and 6.6  $\mu\text{g m}^{-3}$ . The PM<sub>2.5</sub> data were well-within possible future limit values proposed by the European Commission Clean Air For Europe programme. Daily PM<sub>2.5</sub> and PM<sub>10</sub> were significantly correlated ( $r^2 = 0.75$ ) with PM<sub>2.5</sub> contributing 54 %, on average, to PM<sub>10</sub>. The daily BS:PM<sub>10</sub> and BS:PM<sub>2.5</sub> ratios were more variable, and significantly lower in summer than in winter, reflecting the greater contribution of non-black photochemical secondary particles to PM<sub>10</sub> in summer. Analysis with respect to wind showed a dominant influence of dispersion on BS and PM<sub>2.5</sub> but both a dispersion and a wind-driven suspension influence on PM<sub>coarse</sub>. The latter was higher than in central England (averaging about one-third of the PM<sub>coarse</sub>), and greater for on-shore wind direction, suggesting a sea-salt source for this component in addition to other particle resuspension contributions. Overall, the data showed that excursions in PM<sub>10</sub> were driven more by variations in PM<sub>2.5</sub> than by PM<sub>coarse</sub> or BS. Both PM<sub>2.5</sub> and its proportion to PM<sub>10</sub> were significantly elevated for air-masses passing over continental Europe and the British Isles, whereas BS varied less with air-mass origin, supporting the conclusion that concentrations of particulate matter, particularly of finer PM, are strongly influenced by regional scale synoptic meteorology (presumed to be predominantly secondary PM), whereas BS is dominated more by local sources. Comparison of BS with a nearby rural site suggested that approximately three-quarters, on average, of the urban BS was local in origin.

## Introduction

The concentration of outdoor airborne particulate matter is nowadays principally quantified using the  $PM_{10}$  metric. Thus, the European first Daughter Directive (99/30/EC), as for legislation elsewhere, sets limits for  $PM_{10}$  in recognition of the effects of particles on human health.  $PM_{10}$  is of course not a single entity, but comprises a wide range of particle sizes and chemical composition (as a consequence of the multitude of sources and dispersal processes), and it is becoming common to sub-divide  $PM_{10}$  into fine ( $PM_{2.5}$ ) and coarse ( $PM_{10-2.5}$  or  $PM_{coarse}$ ) size ranges. Using this dichotomy it is possible to make the crude generalisation that  $PM_{2.5}$  (which is derived mainly from gas-to-particle reactions in combustion exhaust or between ammonia and sulphate and nitrate) is predominantly of direct anthropogenic origin, while  $PM_{coarse}$  (which is derived mainly from mechanical suspension of soil, dust, sea-salt and diffuse industrial/traffic-related sources) is predominantly of natural or indirect anthropogenic origin (APEG, 1999; AQEG, 2005).

The increasing availability of  $PM_{2.5}$  data for epidemiological studies, principally in the USA, has focused attention on the extent to which this finer fraction of airborne particles is responsible for the health effects previously ascribed to  $PM_{10}$ . Following advice from the World Health Organisation (WHO, 2003; WHO, 2004), the Clean Air for Europe Working Group on Particulate Matter recently recommended that  $PM_{2.5}$  rather than  $PM_{10}$  should become the principal metric for assessing exposure to PM in Europe (CAFE, 2004). However, the CAFE working group has not yet recommended specific limit values for  $PM_{2.5}$ , stating that more European data are required for  $PM_{2.5}$ , and for the factors determining relationships between  $PM_{2.5}$  and  $PM_{10}$  (or  $PM_{coarse}$ ), in order to derive an appropriate legislative framework for the protection of human health against PM. A summary of available  $PM_{2.5}$  monitoring data in Europe has recently been published (Van Dingenen *et al.*, 2004), but caution is required in deriving comparative conclusions because of the variety of sampling and analytical techniques used across the different locations.

An alternative, historic measure of airborne PM is black smoke (BS). This is effectively a measurement of the optical absorption (or blackness) of the PM and can be converted into a notional mass concentration using a standard equation (DETR, 1999). Although the BS sampler

size-selects at approximately  $PM_4$  (McFarland *et al.*, 1982), the optical darkness is dominated by the contribution from sub- $\mu m$  diameter elemental carbon particles (Edwards *et al.*, 1983; Horvath, 1996). Therefore, in the modern-day context, BS is a good surrogate for primary combustion particles, and is largely insensitive to secondary inorganic aerosol and  $PM_{coarse}$ . This fact, in conjunction with source-oriented epidemiological studies that show associations between adverse health and traffic- and combustion-derived air pollution (Laden *et al.*, 2000; Roemer and van Wijnen, 2001; Hoek *et al.*, 2002), has led WHO also to recommend re-evaluation of BS as a particle pollution metric (WHO, 2003), in parallel to their recommendation on  $PM_{2.5}$ .

The paragraphs above show that  $PM_{2.5}$ ,  $PM_{coarse}$  and BS broadly reflect different source contributions to  $PM_{10}$ . The aim of this study was therefore to analyse a year of collocated daily measurements of BS,  $PM_{2.5}$ , and  $PM_{10}$  (and hence  $PM_{coarse}$  by difference) in order to evaluate the factors influencing the daily relationships between these metrics in an urban area in the UK. The data and inferences will contribute to the ongoing development of policy on airborne PM, and in the interpretation of epidemiologic data associating PM air pollution and adverse health effects.

## Experimental

Sampling was conducted on the roof of Old College, an urban background site, in central Edinburgh ( $3^\circ 12' W$ ,  $55^\circ 57' N$ ) for one year from September 1999. The city of Edinburgh (population ~500,000) is situated near the east coast of Scotland, and has comparatively little heavy industry.

Samples of  $PM_{10}$  and  $PM_{2.5}$  were collected daily (midnight to midnight) using R&P Partisol 2025 samplers, both with  $PM_{10}$  heads and one with a sharp cut cyclone to select further for  $PM_{2.5}$ . Samples for BS were collected using a standard UK Black Smoke network sampler. Hourly wind velocity data were logged from a collocated RM Young wind vane. A second Black Smoke sampler was located in a portacabin in a field adjacent to the Centre for Ecology and Hydrology, a rural site 17 km SSW of Edinburgh. The Partisols and BS sampler were operated in accordance with guidance specified by the USEPA (1998) and the UK national smoke network (DETR, 1999), respectively. Every 4 weeks the volumetric flow rate of the Partisols was calibrated

against a NIST traceable critical orifice flow meter (Chinook Engineering). Trials of the two Partisol instruments operated both as PM<sub>10</sub> samplers for 14 days, or both as PM<sub>2.5</sub> samplers for 14 days, yielded correlation coefficients between sample masses of 0.991 and 0.985, respectively.

The Gelman Zefluor filters used in the Partisol samplers were conditioned for 24 h in the weighing room at  $17 \pm 3$  °C and  $53 \pm 7\%$  RH pre- and post-exposure before being passed under an anti-static ionising blower and triple-weighed on a Sartorius MC5 six-place balance. A set of six weigh-blank filters was interspersed within the pre- and post-weight sessions of each batch of sample filters and the mean change in weigh-blank filter mass between weighing sessions used to correct the sample filter mass changes. The mean weigh-blank filter correction corresponded to only 1.1% and 2.1%, respectively, of the mean PM<sub>10</sub> and PM<sub>2.5</sub> sample masses. Green *et al.* (2001) also concluded that there was no significant variability in Partisol sample filters for RH in the range 40 to 60% and temperature in the range 15 to 25 °C.

The reflectance of the Whatman no. 1 filters used in the BS samplers was measured using an EEL Model 43D reflectometer and converted to a mass concentration using the UK BS calibration curve (DETR, 1999). Although this conversion is unlikely accurately to represent true mass concentration of dark particles, it nevertheless provides a relative measure of their concentration.

Air-mass back-trajectories for a mid-day arrival at the 930 hPa pressure level at the sampling site were calculated using the 3-D model available at the British Atmospheric Data Centre. These were categorised by statistical hierarchical clustering using mean Euclidian distance squared and average linkage clustering on the vectors of the 30 variables: distance north of Edinburgh, distance south of Edinburgh, and pressure level, at 12, 24, 36,...120 h prior to arrival. The “best” number of clusters into which to classify the trajectories was adjudged from step-changes in the values of RMS and  $r^2$  plotted against number of clusters (Cape *et al.*, 2000), and examination of the robustness of allocation of individual trajectories to a given cluster. Five major clusters emerged (amalgamating the two Arctic clusters of Heal *et al.* (2005)), together accounting for >94 % of the total number of trajectories.

## **Results and discussion**

## Concentrations

Mean daily  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  and BS for the year of collocated urban measurement were 15.5, 8.5 and  $6.6 \mu\text{g m}^{-3}$ , respectively. Mean daily BS at the rural site was  $1.8 \mu\text{g m}^{-3}$ . The EU Stage 1 limit values (99/30/EC) for  $\text{PM}_{10}$ , incorporated as statutory Air Quality Standard (AQS) objectives in the UK, are an annual mean of  $40 \mu\text{g m}^{-3}$  and no more than 35 days per year (approximately equivalent to 90<sup>th</sup> percentile) of daily means exceeding  $50 \mu\text{g m}^{-3}$ . Proposed tighter AQS objectives (to be achieved by the end of 2010) are no more than 7 exceedences per year (~98<sup>th</sup> percentile) of a daily mean of  $50 \mu\text{g m}^{-3}$  and an annual mean of  $20 \mu\text{g m}^{-3}$  ( $18 \mu\text{g m}^{-3}$  in Scotland). In this study there were no days on which daily  $\text{PM}_{10}$  exceeded  $50 \mu\text{g m}^{-3}$ . This, and the annual mean of  $15.5 \mu\text{g m}^{-3}$ , show that background locations in Edinburgh are already in compliance with the 2010  $\text{PM}_{10}$  objectives.

$\text{PM}_{10}$  was also recorded over the same period by Tapered Element Oscillating Microbalance (TEOM) at an urban centre location in central Edinburgh, ~0.7 km from the Old College site. Mean  $\text{PM}_{10}$  recorded at the TEOM site over the same period was  $17.8 \mu\text{g m}^{-3}$ , also with no daily value exceeding  $50 \mu\text{g m}^{-3}$ . Applying the current “gravimetric equivalent” correction factor of 1.3 to the data to allow for loss of semi-volatile PM in the heated inlet of the TEOM (AQEG, 2005) yielded four days with  $\text{PM}_{10} > 50 \mu\text{g m}^{-3}$ , and an annual average of  $23.1 \mu\text{g m}^{-3}$ . The latter exceeded the proposed 2010 annual AQS, indicating it may be difficult to achieve this more stringent AQS for urban centre locations as well as at specific roadside hotspots, even in a relatively unpolluted city such as Edinburgh.

Correlation of daily  $\text{PM}_{10}$  at the two locations in Edinburgh was fairly high ( $r^2 = 0.53$ ) suggesting that a significant proportion of variability in (non-roadside) urban  $\text{PM}_{10}$  is determined by regional-scale, or background, sources and synoptic meteorology. High local windspeed was the only parameter associated with lack of spatial correlation which is probably due to different characteristics of very localised wind-induced particle suspension in the vicinity of each monitor.

There are no AQSs for  $\text{PM}_{2.5}$  in the UK or elsewhere in Europe. The CAFE Working Group on Particulate Matter suggested  $35 \mu\text{g m}^{-3}$  as a 90<sup>th</sup> percentile daily limit value, and between 12-20

$\mu\text{g m}^{-3}$  as an annual value as starting points for further discussion (CAFE, 2004). For the measurements undertaken here, the 90<sup>th</sup> (and 98<sup>th</sup>) percentile of daily mean  $\text{PM}_{2.5}$  was 15.3 (21.1)  $\mu\text{g m}^{-3}$ , which together with the annual average 8.5  $\mu\text{g m}^{-3}$ , indicate that background air in Edinburgh would also be in compliance with potential future  $\text{PM}_{2.5}$  legislation. The annual mean  $\text{PM}_{2.5}$  in Edinburgh was considerably lower than the  $\text{PM}_{2.5}$  annual means of  $>15 \mu\text{g m}^{-3}$  reported for six other urban background locations in Europe, but are comparable to those summarised for the rural sites investigated (Van Dingenen *et al.*, 2004).

### **Inferences of sources from relationships between PM metrics**

Daily  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  were strongly correlated ( $r^2 = 0.75$ ,  $n = 349$ ), Fig. 1, with linear regression equation,  $\text{PM}_{2.5} = 0.61\text{PM}_{10} - 0.98$ . This is consistent with the strong relationships between hourly  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  from the previous analysis of collocated  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  in the UK (Harrison *et al.*, 2001) which reported  $r^2$  values from 0.60-0.96 and regression coefficients from 0.50-0.80. The recent summary of other European data also reports strong within-site correlation of  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  (Van Dingenen *et al.*, 2004). Since  $\text{PM}_{2.5}$  is a subset of  $\text{PM}_{10}$  some correlation is inevitable, but the extent to which they are correlated is relevant to an assessment of the usefulness of designating an additional standard for  $\text{PM}_{2.5}$ .

The distribution of daily ratios of  $\text{PM}_{2.5}:\text{PM}_{10}$ ,  $\text{BS}:\text{PM}_{10}$  and  $\text{BS}:\text{PM}_{2.5}$ , divided into summer and winter, are shown in Fig. 2. The median daily ratio of  $\text{PM}_{2.5}:\text{PM}_{10}$  for the year was 0.52 (Inter-Quartile Range, IQR: 0.44-0.62). The narrow IQR is a consequence of the fairly strong correlation between  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ . The average proportion of  $\text{PM}_{2.5}$  in  $\text{PM}_{10}$  in Edinburgh ( $\sim 0.55$ ) is towards the low end of the ranges summarised by Harrison *et al.* (2001) for sites in central and southern England and by Van Dingenen *et al.* (2004) for a range of sites across Europe, although comparable to the proportion reported by Brook *et al.* (1997) for sites across Canada. Edinburgh is a comparatively unpolluted city with a windy coastal location, so is likely to be proportionally less influenced by anthropogenic fine particles and more influenced by suspension of coarser dust and sea-salt.

The median (and IQRs) for daily ratios of  $\text{BS}:\text{PM}_{10}$  and  $\text{BS}:\text{PM}_{2.5}$  were 0.42 (0.27-0.59) ( $r^2 = 0.18$ ,  $n = 349$ ) and 0.80 (0.51-1.09) ( $r^2 = 0.27$ ,  $n = 362$ ), respectively. The poorer correlation and



much wider variability in these latter ratios compared with  $PM_{2.5}:PM_{10}$  data reflects the fact that black smoke is a strong indicator for a specific source contributor to PM (combustion-derived dark particles) rather than being a measure of all particles in a given size fraction. Fig. 2 also shows that, on average, the ratios  $BS:PM_{10}$ , and  $BS:PM_{2.5}$ , were significantly lower ( $P < 0.001$ ) in summer than in winter, reflecting either or both greater contribution of non-black photochemical secondary particles to PM in summer and increased BS from combustion heating in winter. There are no local sources of BS at the rural site, so from the average rural:urban BS ratio of ~0.25 it can be inferred that nearly three-quarters of BS in Edinburgh is locally generated and the remainder is “background.” (The above analysis excluded days when winds were from the N-NE for which Edinburgh is immediately upwind of the rural site. A dispersion source from the city was very clear on these occasions, with rural:urban BS ratios exceeding 0.5).

Fig. 1 reveals slight upward curvature with increasing particle concentration. There was a significant trend for the contribution of  $PM_{2.5}$  to  $PM_{10}$  to increase with increasing  $PM_{2.5}$  (Fig. 3) and, less strongly, with increasing  $PM_{10}$ . In addition there was greater variation in daily  $PM_{2.5}$  than in  $PM_{coarse}$  (90%ile-median values of 8.0 and 5.4  $\mu g\ m^{-3}$  for  $PM_{2.5}$  and  $PM_{coarse}$ , respectively) and little correlation between  $PM_{2.5}$  and  $PM_{coarse}$  ( $r^2 = 0.10$ ). All this can be interpreted as showing that days of high  $PM_{2.5}$  were not also strongly associated with high  $PM_{coarse}$  and that variability in daily  $PM_{10}$  was more driven by variations in  $PM_{2.5}$  than in  $PM_{coarse}$ . Van Dingenen *et al.* (2004) also report a tendency for  $PM_{2.5}:PM_{10}$  ratio to be lower at sites with lower  $PM_{10}$  concentration, except at heavily trafficked kerbside sites where traffic-induced suspension of coarse particles make a substantial contribution to local  $PM_{10}$ .

Further insight is gained from analysis of PM size fractions with local windspeed.  $PM_{10}$  and BS are plotted against daily average windspeed in Fig. 4 and show clearly distinct relationships. The relationship of  $PM_{10}$  with windspeed is scattered but has a U-shape showing levels of  $PM_{10}$  to be enhanced at both high and low windspeeds. The observation is consistent with the action of the opposing processes of entrainment of  $PM_{10}$  (or components of  $PM_{10}$ ) with increasing windspeed on the one hand, and dilution by wind on the other. The latter process is illustrated well in Fig. 4 by the strongly monotonic inverse relationship between windspeed and BS, which consists almost entirely of fine particles emitted from local combustion sources. The  $PM_{2.5}$  data has a

similar relationship to windspeed as BS, indicating that it is the  $PM_{coarse}$  component of  $PM_{10}$  which includes a windspeed-dependent resuspension source. This source will be wind-driven entrainment of dust on soils and other surfaces. For a coastal location like Edinburgh, this component is likely also to include sea-salt, whose contribution to atmospheric PM will increase with increasing windspeed over the sea surface.

The effect of dispersion dilution on  $PM_{coarse}$  can be factored out by normalising with respect to the BS data, as shown in Fig. 5. Despite the scatter in the data there is a significant trend for  $PM_{coarse}/BS$  to increase with windspeed, and more strongly in the summer than in the winter. The seasonal difference may be explained by generally drier conditions in summer leading to more facile suspension of particles from the surface, although closer inspection of Fig. 5 shows that highest windspeeds more often occurred in winter.

Using the methodology described by Harrison *et al.* (2001), power-law relationships between windspeed and resuspended  $PM_{coarse}$  were derived for each season, from which the overall magnitude contributed by resuspended  $PM_{coarse}$  could be estimated. A threshold windspeed for resuspension of  $2 \text{ m s}^{-1}$  best fit both the summer and winter data. During summer, resuspended  $PM_{coarse}$  contributed  $1.9 \mu\text{g m}^{-3}$  (or 26 %) on average to mean total  $PM_{coarse}$  ( $7.1 \mu\text{g m}^{-3}$ ), whilst in winter its estimated contribution was  $2.6 \mu\text{g m}^{-3}$  (or 37 %). Thus, while the data show that resuspension is more facile in summer, the higher average windspeed during winter actually results in a larger component of resuspended  $PM_{coarse}$  in  $PM_{10}$  during winter. The proportions of resuspended  $PM_{coarse}$  derived here for Edinburgh are generally higher than those determined by Harrison *et al.* (2001) for Birmingham, central London and Harwell because of Edinburgh's windier and near-coastal location. This more detailed analysis concurs with the earlier observation that  $PM_{coarse}$  comprises a greater proportion of  $PM_{10}$ , on average, in Edinburgh, than at the sites investigated by Harrison *et al.* (2001).

The remaining non-wind-suspended component of  $PM_{coarse}$  will include primary emissions of coarse particles from industry and construction activity, as well as the important source of traffic-related coarse particles arising via mechanical abrasion from the vehicle and entrainment from the road surface by vehicle-induced turbulence. Analysis of chemical composition data has also

demonstrated that non-exhaust vehicle emissions make an important contribution to urban PM in the UK (Harrison *et al.*, 2004; Heal *et al.*, 2005).

A contribution to  $PM_{coarse}$  from sea-salt is supported by the wind roses plotted in Fig. 6 which show that  $PM_{coarse}$  is elevated when local wind direction is from the north-east and east. This is the coastal sector from central Edinburgh, and a windspeed wind rose (not shown) shows that windspeeds from this sector are higher than average. Fig. 6 also shows that  $PM_{2.5}$  is proportionally more enhanced than  $PM_{coarse}$  when wind is from the east and this must be due to longer-range transport of particles from continental Europe. The significant influence of long-range transport on concentrations of  $PM_{10}$  is well-known (King and Dorling, 1997; Malcolm *et al.*, 2000; Buchanan *et al.*, 2002). Here the influence of long-range transport on different particle measures was investigated according to the trajectory taken by the air-mass arriving at the measurement site. Fig. 7 shows that median  $PM_{10}$  on days when air-mass originated from continental Europe or from local circulation about the British Isles ( $20$  and  $16 \mu g m^{-3}$ , respectively) were significantly elevated compared with days when air-mass originated from Atlantic SW, Atlantic W and the Arctic ( $12$ ,  $13$  and  $14 \mu g m^{-3}$ , respectively). The pattern was accentuated for  $PM_{2.5}$  with median concentrations of  $11$  and  $9 \mu g m^{-3}$  for days with air-masses originating from continental Europe or local circulation (i.e. over land) compared with  $6 \mu g m^{-3}$  when air-masses originated from the Atlantic and Arctic. Thus change in air-mass source was associated with increases of 50 % or more, on average, of receptor  $PM_{2.5}$  in Edinburgh. The ratio of  $PM_{2.5}/PM_{10}$  was also significantly elevated on the days with European and local circulation air-masses as shown by the numbers at the bottom of Fig. 7.

In contrast, BS in Edinburgh varied much less with air-mass source than  $PM_{10}$  or  $PM_{2.5}$ . The max/min of the median trajectory cluster values shown in Fig. 7 are 1.7 and 1.9 for  $PM_{10}$  and  $PM_{2.5}$ , respectively, but only  $\sim 1.4$  for BS, implying that urban BS is more dominated by local sources and less impacted by the air-mass origin than other components of  $PM_{10}$ . Consequently there is a lower urban BS: $PM_{10}$  ratio for polluted European trajectories. (Fig. 7 shows BS to be second highest on average for the Arctic trajectories but this is, in part, due to the urban monitoring site being on the south side of the city centre. The key point is the lower variability of BS with air mass source region than for  $PM_{2.5}$  and  $PM_{10}$ ). Rural BS was also elevated for the

European and local circulation air-mass back trajectories, but the differences amounted only to  $\sim 1 \mu\text{g m}^{-3}$  in absolute concentration (data not plotted). Thus, although there exists a “background” component of BS influenced by long-range transport, it is a minor source of BS compared with BS generated in the local vicinity of the urban area, as highlighted above from consideration of urban:rural BS ratios. These analyses have yielded more detailed support to the earlier conclusion of Buchanan *et al.* (2002) that BS is these days rather less influenced by long-range transport than  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ .

## Conclusions

European data for collocated  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  remain fairly scarce despite strong moves towards defining  $\text{PM}_{2.5}$  air quality objectives for protection of human health. The combination of PM and black smoke measurements can provide insight into controlling influences on receptor particle concentrations. This work has shown that  $\text{PM}_{2.5}$  in background air in a medium-sized UK city is likely to be within potential future air quality objectives, and that, on average,  $\text{PM}_{2.5}$  is strongly correlated with  $\text{PM}_{10}$ . If chronic exposure to particles in the long-term (years) dominates the adverse health burden then measurement of one metric in a locality may suffice. On the other hand if short-term (daily) variability in exposure to particles is important then this has been shown to be driven more by variation in  $\text{PM}_{2.5}$ , than by  $\text{PM}_{\text{coarse}}$  or by combustion-related particles as characterised by black smoke. Either way, variation in PM is strongly influenced by local and synoptic meteorology outwith local control measures, with combustion-related dark particles and coarse particles related more to local sources dependent on dispersion and suspension, and finer particles related more to regional scale transport. By understanding the variance of the sources and determinants of the particle matter heterogeneous mix it may be possible to offer a better interpretation of the epidemiologic data associating particulate air pollution with adverse health outcomes.

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## Figure Captions

Figure 1: Relationship between daily  $PM_{10}$  and  $PM_{2.5}$  in urban background air in Edinburgh. (Summer, Apr-Sep; Winter, Oct-Mar).

Figure 2: Distribution of ratios between daily values of  $PM_{10}$ ,  $PM_{2.5}$  and black smoke (BS) in urban background air in Edinburgh. (Summer, Apr-Sep; Winter, Oct-Mar).

Figure 3: Relationship of  $PM_{2.5}:PM_{10}$  ratio against  $PM_{2.5}$  for daily measurements in urban background air in Edinburgh.

Figure 4: Relationship between daily  $PM_{10}$  and black smoke and local windspeed in Edinburgh. (Summer, Apr-Sep; Winter, Oct-Mar).

Figure 5: Ratio of  $PM_{coarse}/BS$  with windspeed in Edinburgh, and best-fit lines. (Summer, Apr-Sep; Winter, Oct-Mar).

Fig. 6: Wind roses of collocated  $PM_{10}$ ,  $PM_{2.5}$ ,  $PM_{coarse}$  and BS in background air in Edinburgh. Data points are moving averages of three consecutive  $10^\circ$  wind-sector values.

Fig 7: Apportionment of daily  $PM_{10}$  and  $PM_{2.5}$ , and  $PM_{2.5}/PM_{10}$  ratio, in Edinburgh by geographical sector of 5-day air-mass back-trajectory. Clusters are ordered in descending median  $PM_{10}$  value.

Fig. 1: Relationship between daily  $PM_{10}$  and  $PM_{2.5}$  in urban background air in Edinburgh.  
(Summer, Apr-Sep; Winter, Oct-Mar).

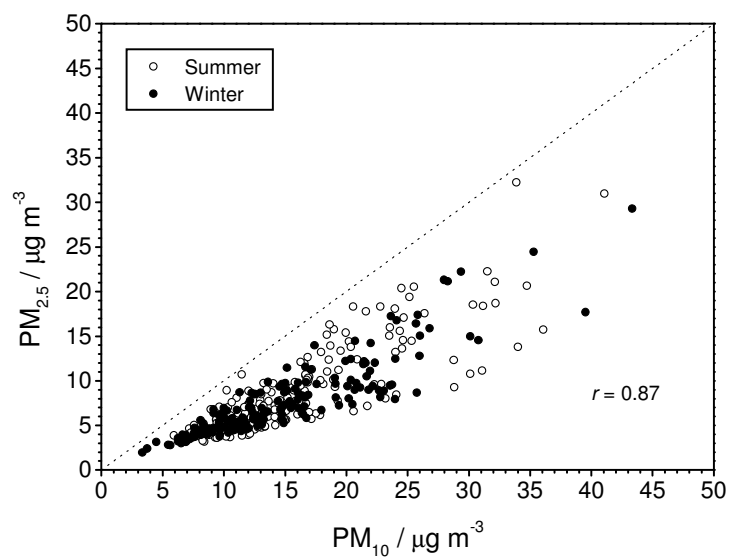




Fig 2: Distribution of ratios between daily values of PM<sub>10</sub>, PM<sub>2.5</sub> and black smoke (BS) in urban background air in Edinburgh. (Summer, Apr-Sep; Winter, Oct-Mar).

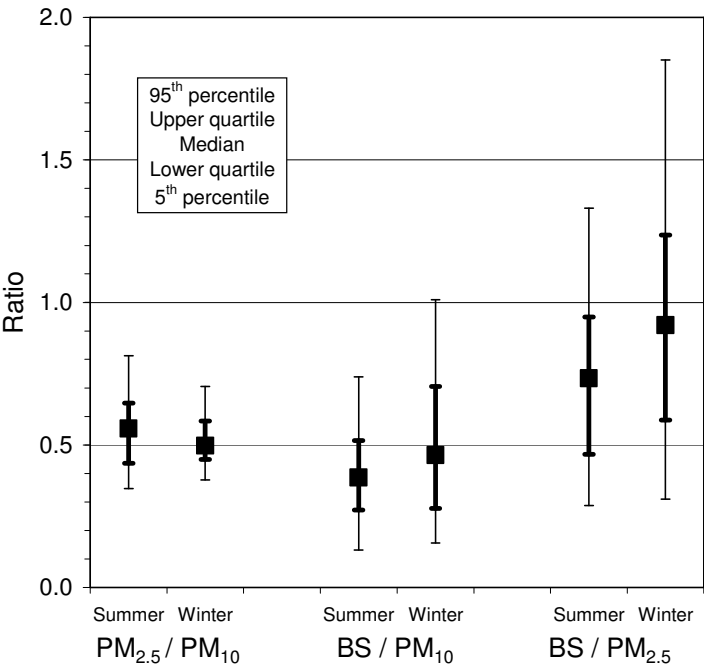


Fig 3: Relationship of  $PM_{2.5}:PM_{10}$  ratio against  $PM_{2.5}$  for daily measurements in urban background air in Edinburgh.

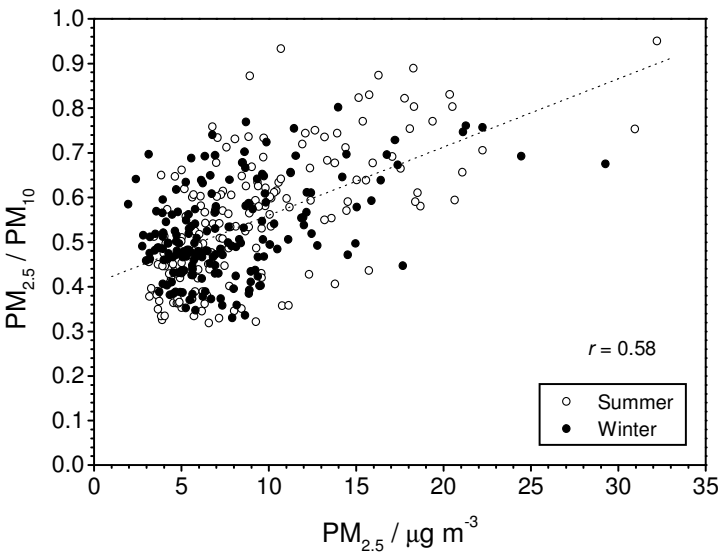


Fig. 4: Relationship between daily  $PM_{10}$  and black smoke and local windspeed in Edinburgh.  
(Summer, Apr-Sep; Winter, Oct-Mar).

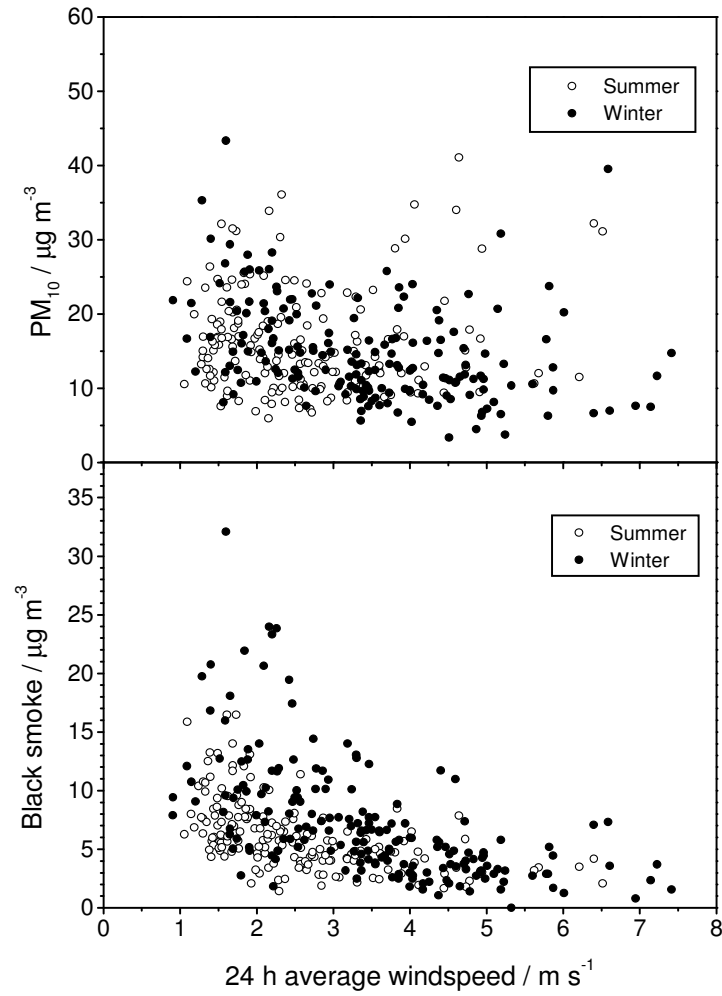


Fig. 5: Ratio of  $\text{PM}_{\text{coarse}}/\text{BS}$  with windspeed in Edinburgh, and best-fit lines. (Summer, Apr-Sep; Winter, Oct-Mar).

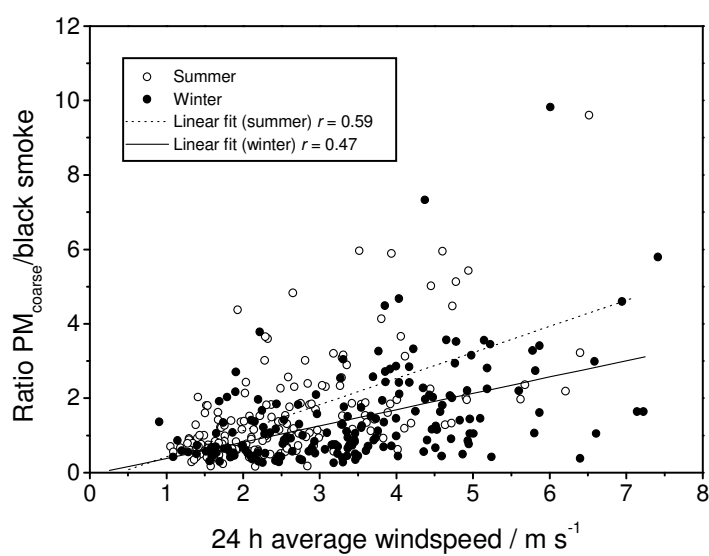


Fig. 6: Wind roses of collocated  $PM_{10}$ ,  $PM_{2.5}$ ,  $PM_{coarse}$  and BS in background air in Edinburgh. Data points are moving averages of three consecutive  $10^\circ$  wind-sector values.

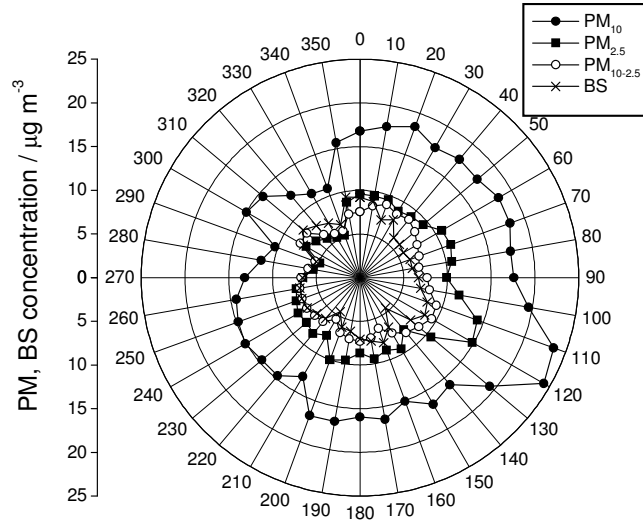


Fig 7: Apportionment of daily  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , and  $\text{PM}_{2.5}/\text{PM}_{10}$  ratio, in Edinburgh by geographical sector of 5-day air-mass back-trajectory. Clusters are ordered in descending median  $\text{PM}_{10}$  value. The number of trajectories in each cluster is given in the axis labels.

